1	Intercomparison of Aerosol Optical Depths from four reanalyses and their
2	multi-reanalysis-consensus
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21	Key Points:
22 23 24 25	<ol> <li>Four global aerosol reanalyses are intercompared and verified with observations for their skill in simulating aerosol optical depth.</li> <li>The study identifies the strength of each reanalysis and the regions where there are notable differences and challenges.</li> </ol>
25 26	3. The multi-reanalysis-consensus, based on the four reanalyses, consistently ranks as one
27	of the best regionally and globally.
28	
29	Abstract
30	The emergence of aerosol reanalyses in recent years has facilitated a comprehensive and
31	systematic evaluation of Aerosol Optical Depth (AOD) trends and attribution over multi-decadal
32	timescales. Notable <u>multiyear</u> aerosol reanalyses currently available include NAAPS-RA from the
33 34	U.S. Naval Research Laboratory; the NASA MERRA-2; JRAero from the Japan Meteorological Agency (JMA); and CAMSRA from Copernicus/ECMWF. These aerosol reanalyses are based on
34 35	differing underlying meteorology models, representations of aerosol processes, and data
35 36	assimilation methods and treatment of AOD observations. This study presents the basic
37	verification characteristics of these four reanalyses versus both AERONET and MODIS retrievals
38	in monthly AOD properties and identifies the strength of each reanalysis and the regions where

diversitydivergence and challenges are prominent. Regions with high pollution and often mixed

fine-coarse mode aerosol environments such as South Asia, East Asia, Southeast Asia, and the

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Maritime Continent pose significant challenges, as indicated by higher monthly AOD root mean 41 square error. Moreover, regions that are distant from major aerosol source areas, including the 42 polar regions, and remote oceans, exhibit large relative differences in speciated AODs and fine-43 mode vs coarse-mode AODs among the four reanalyses. To ensure consistency across the globe, 44 45 a multi-reanalysis-consensus (MRC. i.e. ensemble mean) approach was developed similar to the International Cooperative for Aerosol Prediction Multi-Model Ensemble (ICAP-MME). Like the 46 47 ICAP-MME, while the MRC does not consistently rank first among the reanalyses for individual regions, it performs well by ranking first or second globally in AOD correlation and RMSE, 48 49 making it a suitable candidate for climate studies that require robust and consistent assessments.

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51 Keywords: Aerosol, Reanalysis, Aerosol Optical Depth, intercomparison, ICAP-MME

- 52
- 53 Short Summary

The study compares and evaluates monthly aerosol optical depth of four reanalyses (RA) and their consensus- (i.e. ensemble mean). The basic verification characteristics of these RA versus both

consensus-<u>(i.e. ensemble mean).</u> The basic verification characteristics of these RA versus both AERONET and MODIS retrievals are presented. The study discusses the strength of each RA and

identifies regions where diversity divergence and challenges are prominent. The RA consensus

usually performs very well on a global scale in terms of how well it matches the observational

59 data, making it a good choice for various applications.

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#### 61 1. Introduction

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In recent years, global aerosol reanalyses have been developed by major operational and research 62 centers, owing to the availability of long-record satellite remote sensing aerosol products and 63 advancements in aerosol data assimilation and modeling. These reanalyses are based on their 64 operational counterparts that are included in the "Core Four" members of the International 65 Cooperative for Aerosol Prediction Multi Model Ensemble (ICAP-MME C4C; Sessions et al., 66 67 20162015; Xian et al., 2019; Reid et al., 2022). The reanalyses include the Copernicus Atmosphere 68 Monitoring Service ReAnalysis (CAMSRA; Inness et al., 2019) produced by the European Centre 69 for Medium-Range Weather Forecasts (ECMWF); the Japanese Reanalysis for Aerosol (JRAero) (Yumimoto et al., 2017) developed by the Japan Meteorological Agency (JMA); the NASA 70 71 Modern-Era Retrospective Analysis for Research and Applications, version 2 (MERRA-2; 72 Randles et al., 2017); and the Navy Aerosol Analysis and Prediction System reanalysis (NAAPS-73 RA; Lynch et al., 2016) developed by the U.S. Naval Research Laboratory (NRL).

74 The aerosol reanalyses are similar to their operational counterparts and characterized by a high 75 degree of independence in their underlying meteorology, aerosol sources, sinks, microphysics, and 76 chemistry, as well as in their assimilation methods for aerosol optical depth (AOD) observations. 77 A summary of the configurations of these four reanalyses is presented in Table 1 for general 78 features and Table 2 for microphysical and optical treatments of different aerosol species. Notably, 79 the use of operational Terra and Aqua Moderate Resolution Imaging Spectrometer data (MODIS 80 Dark Target and Deep Blue; Levy et al., 2013; Hsu et al., 2013) is consistent across these 81 reanalyses, although preprocessing treatments vary. These treatments include quality control, bias 82 correction, and aggregation and sampling. Additionally, several other products, such as MultiAngle Imaging Spectroradiometer (MISR; Kahn et al., 2010), Advanced Very High 83 Resolution Radiometer (AVHRR; e.g., Ignatov et al., 2002), and Polar Multi Sensor Aerosol 84 85 product (PMAp; GrzegorskiAdvanced Along-Track Scanning Radiometer (AATSR, Popp et al., 2022),2016) are assimilated into some of these reanalyses, although these additional remote 86 87 sensing data probably have only a small impact during the MODIS era, as their data volume is 88 small compared to MODIS. Therefore, between their underlying meteorology, physics, and data

assimilation these reanalyses are characterized by a high degree of independence overall.

Like atmospheric reanalysis products, aerosol reanalysis products, whether used individually or in 90 combination, have been employed for diverse applications. They provide comprehensive aerosol 91 climatology and statistics to aid in understanding aerosol conditions across various regions and the 92 world (e.g., Reid et al., 2012; Xian et al., 2020; Nignombam et al., 2021; Ohno et al., 2022; Rubin 93 94 et al., 2023). They are widely used to address a multitude of scientific inquiries in the fields of aerosol radiative forcing (e.g., Randles et al., 2017; Markowicz et al., 2017; 2021a,b; Ohno et al., 95 2022; Zhang et al., 2023), aerosol-cloud interaction (e.g., McCoy et al., 2017; Ross et al., 2018; 96 Eck et al., 2018), aerosol-cryosphere interaction (e.g., Khan et al., 2018, 2019; 2020; 97 Roychoudhury et al., 2022), air quality and its impact on health (e.g., Tong et al., 2023; Cui et al., 98 99 2022; Jenwitheesuk et al., 2022; Lacima et al., 2022), biogeochemical cycles (e.g., Rahav et al., 2020; Borchardt et al., 2019; Mescioglu et al., 2019), among others. These reanalyses have been 100 rigorously evaluated by the developing centers and various studies from different perspectives, 101 including AOD and other aerosol optical properties, mass concentrations, and vertical distribution 102

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profiles. However, to date, no intercomparison among the four reanalyses has been conducted.

104 This study presents an intercomparison of the four available global aerosol reanalyses to evaluate their skill in simulating monthly average AOD. Additionally, this study includes the development 105 of a Multi Reanalysis Consensus (MRC) product using a multi-model-consensus approach, similar 106 to the ICAP Multi Model Ensemble (ICAP-MME; Sessions et al., 2015; Xian et al., 2019). The 107 MRC is a consensus an ensemble mean (i.e., mathematical average) of the four individual 108 reanalyses, with a spatial resolution of 1°x1° latitude/longitude and monthly temporal resolution. 109 The study provides speciated AODs, fine-mode (FM), coarse-mode (CM) and total AODs at 550 110 111 nm for the period of 2003-2019 from three reanalyses, and all four reanalyses are available for the 112 time period of 2011-2019. In addition, a companion study focuses on global and regional AOD 113 trends derived from these reanalyses. The validation of AODs from the MRC, and the four component members, is performed using ground-based AEROsol Robotic NETwork (AERONET; 114 Holben et al., 1998) observations, with MODIS AOD for spatial distribution evaluation. The 115 116 validation results, as well as the AOD climatology and diversitydivergence of the reanalyses, are presented in Section 3. The study concludes with a summary of the findings in Section 4. 117

# 118119 2. Data and Methods

This study intercompares the monthly average modal (total, FM, and CM) and speciated AOD
products from four aerosol reanalyses (RA) and their consensus, and evaluates the RA AODs with
AERONET and the combined MODIS Dark Target/Deep Blue retrievals (Levy et al., 2013; Hsu
et al., 2013).

- 124 2.1 Individual product lines

125 Descriptions of the four reanalysis datasets, including CAMSRA, JRAero, MERRA-2, and 126 NAAPS-RA v1, are provided in this section. Table 1 provides a summary of the features of the 127 four reanalyses and the MRC used in this study. basic features of the four reanalyses and the MRC used in this study. Table 2 offers a summary of the parameters employed to depict the 128 microphysical and optical properties of aerosol species from these reanalyses. Furthermore, Table 129 130 3 samples hygroscopic enhancement factor values that influence optical property calculations due 131 to the hygroscopic growth of particles at various relative humidity levels. In addition to utilizing 132 different meteorological data, aerosol source data, AOD observations, and constructing aerosol 133 species, notable differences exist even among similar species regarding treatments related to 134 aerosol microphysics, optical properties, and water uptake ability for hydrophilic species.

135 2.1.1 CAMSRA

136 The Copernicus Atmosphere Monitoring Service (CAMS) Reanalysis (CAMSRA, Inness et al.,

137 2019) is run at the European Centre for Medium-Range Weather Forecasts (ECMWF) and is a

138 global reanalysis of atmospheric composition species, including aerosols. It builds on the

previous reanalyses of the MACC project (Inness et al., 2013) and the CAMS interim reanalysis
(Flemming et al., 2017). The CAMSRA is publicly available for the years 2003 to 2022 and is

(Flemming et al., 2017). The CAMSRA is publiclybeing continuously updated for future years.

142 The CAMSRA is based on the Integrated Forecasting System (IFS) used by ECMWF for numerical weather prediction and meteorological reanalysis. Two additional modules are 143 incorporated into the IFS for the CAMSRA, one to calculate the processes and reactions of the 144 chemical species and one to represent the prognostic aerosol species. The aerosol scheme 145 includes prescribed and online emissions, dry and wet deposition, production of sulfate from a 146 147 gas-phase sulfur dioxide precursor, and the aging of hydrophobic organic matter (OM) and black 148 carbon (BC) to hydrophilic. The prescribed anthropogenic emissions come from the MACCity 149 inventory (Granier et al., 2011) and the biomass burning (BB) emissions from the Global Fire 150 Assimilation System, version 1.2 (GFASv1.2) (Kaiser et al., 2012). GFASv1.2 is a separate 151 system to the IFS that uses satellite retrievals of fire radiative power to produce the BB emissions that are then input as fixed emissions to the aerosol scheme. The transport of the aerosol species 152 by advection, convection and diffusion is calculated using the meteorological component of the 153 154 IFS and the wind fields from the meteorology are also used as parameters to estimate the online sea salt (Monahan et al., 1986) and dust (Ginoux et al., 2001) surface emissions. One key 155 difference between the CAMSRA set up of the IFS and that used for numerical weather 156 prediction, is that for the CAMSRA the radiative impact of aerosol particles and ozone on 157

158 meteorology is also accounted for.

159 The observations used in the CAMSRA for aerosols are of total AOD at 550nm. These come

160 from MODIS collection 6 satellite retrievals for the entire period covered by CAMSRA and from

the Advanced Along-Track Scanning Radiometer for the period 2003-2012. These AOD

162 observations are simultaneously assimilated with trace gas and meteorological observations

using the 4D variational data assimilation system of the IFS with a 12-hour assimilation window.The products available from the CAMSRA include speciated AODs at a 3-hour temporal and

approximately 0.7 degrees spatial resolution, whereas monthly mean AODs at 550nm were used

166 in this study.

#### 167 2.1.2 JRAero

The Japanese Reanalysis for Aerosol (JRAero) was developed by the Meteorological Research Institute (MRI) of the Japan Meteorological Agency and Kyushu University using the global aerosol transport model MASINGAR Mk-2 (Yukimoto et al., 2012) and a two-dimensional variational (2D-Var) data assimilation method. The model uses the MRI-AGCM3 atmospheric general circulation model, and considers major tropospheric aerosol components, including black

173 carbon (BC), organic carbon (OC), mineral dust, sea salt, and sulfate aerosols, and their precursors.

JRAero assimilates global AOD from a bias-corrected MODIS Level 3 AOD product provided by 174 the US Naval Research Laboratory (NRL) and the University of North Dakota 175 (http://doi.org/10.5067/MODIS/MCDAODHD.NRT.061) every 6 hours. Anthropogenic and 176 biomass burning emissions were estimated using the MACCity (MACC/CityZEN EU projects) 177 178 emission inventory (http://accent.aero.jussieu.fr/MACC\_metadata.php) and the Global Fire 179 Assimilation System (GFAS) dataset (http://www.gmes-180 atmosphere.eu/about/project\_structure/input\_data/d\_fire). The reanalysis has a resolution of TL159 (about  $1.1^{\circ} \times 1.1^{\circ}$ ) with 48 vertical layers from the ground to 0.4 hPa. Validation results and additional information can be found in Yumimoto et al. (2017).

#### 183 2.1.3 MERRA-2

184 The NASA Modern-Era Retrospective Analysis for Research and Applications, version 2

(MERRA-2, Gelaro et al. 2017) is an atmospheric and aerosol reanalysis produced with the
 NASA Goddard Earth Observing System (GEOS) Earth system model. Aerosol data assimilation

187 brings in data from the MODIS and MISR satellite sensors (after 2000) and includes AERONET

188 ground-based sun photometer observations (through 2014). The Goddard Chemistry, Aerosol,

189 Radiation, and Transport model (GOCART; Chin et al. 2000; Colarco et al. 2010) is run online

190 and radiatively coupled in the MERRA-2 system, and provides simulations of dust, sea salt,

191 sulfate, and black and organic carbon aerosol species.

Black and organic carbon are each partitioned into hydrophobic and hydrophilic modes, and a 192 single bulk sulfate aerosol species is carried. Dust and sea salt are partitioned into five non-193 interacting size bins, with dust emissions based on the model 10-m wind speed and a topographic 194 source function following Ginoux et al. (2001), and sea salt emissions driven by the surface wind 195 friction speed modified from Gong (2003) and with a sea-surface temperature adjustment based 196 on Jaeglé et al. (2011). Explosive volcanic sulfur emissions are included through 2010 based on 197 Diehl et al. (2012), with a repeating annual cycle of degassing volcanic emissions subsequent. 198 Other emissions are as summarized in Table 1. 199

200 The analysis of AOD is performed on quality-controlled MODIS, MISR, and AERONET data as described in Randles et al. (2017) and Buchard et al. (2015). The AOD analysis is performed by 201 means of analysis splitting, where first a 2-D analysis of AOD is performed using error 202 203 covariances derived from innovation data. Three-dimensional analysis increments for aerosol 204 mass concentration are then computed using the Local Displacement Ensemble (LDE) methodology, which accommodates misplacement of the aerosol plumes due to source or 205 transport issues. The ensemble perturbations are generated at the full model resolution, without 206 207 the need for multiple model runs. Online quality control is performed as in Dee et al. (2001), with observation and background errors estimated as in Dee and da Silva (1999). Randles et al. 208 (2017) and Buchard et al. (2017) describe the overall methodology and validation of the 209 MERRA-2 AOD reanalysis. For this study, monthly mean speciated AODs and total AOD at 550 210 211 nm with 0.5 degree latitude and 0.625 degree longitude spatial resolution were used.

212 2.1.4 NAAPS-RA v1

The Navy Aerosol Analysis and Prediction System (NAAPS, Lynch et al., 2016) is a global offline chemical transport model developed at the U.S. Naval Research Laboratory. NAAPS simulates the life cycles of aerosol particles and their gaseous precursors. The particle species include anthropogenic and biogenic fine (ABF, a mix of sulfate, organic aerosols and BC from non-BB sources), BB smoke, aeolian dust, and sea salt aerosols. The transport, hygroscopic growth of particles, dry and wet removal processes of these particles, and emissions of wind-blown particles are driven by the meteorological fields from the Navy Global Environmental model (NAVGEM, 220 Hogan, et al., 2014). Secondary organic aerosol (SOA) processes are represented with a 1<sup>st</sup> order approximation method, in which production of SOA from its precursors is assumed to be instant 221 and is pre-treated outside the model. Anthropogenic emissions come from the MACC inventory 222 from ECMWF (Granier et al., 2011). BB smoke emission is derived from the Fire Locating and 223 Modeling of Burning Emissions (FLAMBE, Reid et al., 2009), which is constructed based on the 224 225 MODIS fire hot spot data. In the reanalysis version, additional orbital corrections and regional emission factors are incorporated. Aeolian dust emissions are determined based on the surface 226 227 friction velocity to the fourth power, and surface erodibility, which is adopted from Ginoux et al. 228 (2001) with regional tuning. Dust emission occurs when specific conditions related to surface 229 wetness and friction velocity thresholds are met. The representation of sea spray process adheres

to Witek et al. (2007), with sea salt emission being governed by sea surface wind conditions.

231 The NAAPS-ReAnalysis (NAAPS-RA) v1 (Lynch et al., 2016) is derived from NAAPS, with 232 assimilation of quality-assured and quality-controlled MODIS (Zhang et al., 2006; Hyer et al. 2011) and MISR AOD products (Shi et al., 2011) using 2D-variational-var data assimilation 233 234 method (Zhang et al., 2008). It provides 3-D mass concentration, extinction, and 2-D 550 nm AOD from these aerosol species with 1°x1° latitude/longitude spatial and 6-hourly temporal resolution 235 for the years 2003-2022. The BB smoke source and dust sources are regionally tuned to best match 236 the FM and CM AODs with AERONET AODs. Aerosol wet removals within the tropical region 237 were regulated with satellite precipitation product (Xian et al., 2009) to mitigate model's 238 deficiency to simulate convective precipitation. The reanalysis shows similar decadal trend of 239 AOD found in satellite products (e.g., Zhang et al., 2017) and was verified with various field 240 campaign data (e.g., Reid et al., 2016; Atwood et al., 2017; Edwards et al., 2022; Reid et al., 2023) 241 in addition to ground and space-based observations. 242

## 243 2.2 Multi-reanalysis-consensus (MRC)

The MRC product is a result of combining four individual aerosol reanalysis products described 244 above. This method follows the multi-model-ensemble approach used by the International 245 Cooperative for Aerosol Prediction (ICAP) and is based on the work by Sessions et al. (2015) and 246 Xian et al. (2019). The data from each RA with spatial resolution different from 1°x1° lat/lon 247 degree, is first projected onto the global map with 1°x1° lat/lon degree resolution using linear 248 interpolation. Then the MRC value is determined by calculating the average of the values from the 249 250 four RAs. No weighting among the RAs is applied, or the four RAs are weighted equally in 251 deriving MRC. Regionally-weighted ensemble product based on the verification results shown 252 here can be developed in the future. The MRC provides speciated and total AOD at 550 nm with 253 a 1°x1° lat/lon degree - and monthly - resolution for the period 2011-2003-2019. Data The MRC data 254 for the period spanning from 2003- to 2010 are available from all-relies on three individual 255 reanalyses exceptRAs, while for the period from 2011 to 2019, it incorporates all four RAs, 256 considering that JRAero- data is only accessible starting from 2011.

257 Table 1. Summary of the characteristics of the aerosol reanalyses.

	Organization	Meterology	Resolution lat x lon	DA metho	Assimilated obs.	Species	Anthro. & Biogenic Emission	BB Emissions	Available time	reference	
CAMSRA	ECMWF	Inline ERA5	0.7 x 0.7	4D-Var	DAQ MODIS PMAp	BC, OM, Sulfate Dust, Sea Salt	MACCity (trend: ACCMIP + RCP8.5), monthly VOC	GFAS	2003- present	Inness et al., 2019	
				2D-Var	Neural Net MODIS,	BC, OC, Sulfate	EDGAR V4.1,	GFED before 2009,	1980-	Randles et al., 2017	
MERRA-2	NASA	Inline MERRA-2	0.5 x 0.6	+LDE	MISR, AVHRR, AERONET	Dust, Sea Salt	AeroCom Phase II	QFED after 2009	present		
NAAPS-R	A NRL	Offline NOGAPS/NAVGEM	1 x 1	2D-Var	DAQ MODIS, MISR	BB smoke, Dust, Sea Salt, ABF	MACCity, BOND POET, monthly SOA	FLAMBE	2003- present	Lynch et al., 2016	
JRAero	JMA	Inline MRI AGCM3	1.1 x 1.1	2D-Var	DAQ MODIS	BC, OC, Sulfate Dust, Sea Salt	MACCity	GFAS	2011- present	Yumimoto et al., 2017	
MRC	-	-	1 x 1	-		BB smoke, Dust, Sea Salt, ABF	-		2003- present	this work	

	Developer	Meterology	Resolution lat x lon		Assimilated obs.	Species	Anthro. & Biogenic Emission	BB Emissions	Available time	reference	
CAMSRA	ECMWF	Inline ERA5	0.7 x 0.7	4D-Var	DAQ MODIS, AATSR	BC, OM, Sulfate Dust, Sea Salt	MACCity (trend: ACCMIP +RCP8.5), monthly VOC		2003-present	Inness et al., 2019	
		Inline MERRA-2	0.5 x 0.6	2D-Var	Neural Net MODIS,	BC, OC, Sulfate	EDGAR V4.1,	GFED before 2009,			
MERRA-2	NASA			+LDE	MISR, AVHRR, AERONET	Dust, Sea Salt	AeroCom Phase II	QFED after 2009	1980-present	Randles et al., 2017	
NAAPS-RA	NRL	Offline	1 x 1	2D-Var	DAQ MODIS, MISR	BB smoke, Dust,		FLAMBE	2003-present	Lynch et al., 2016	
		NOGAPS/NAVGEM				Sea Salt, ABF	POET, monthly SOA	TENNOL	2000 present	-,,	
JRAero	JMA	Inline MRI AGCM3	1.1 x 1.1	2D-Var	DAQ MODIS	BC, OC, Sulfate	MACCity	GFAS	2011-present	Yumimoto et al., 201	
0101010	0110 (		1.1.4.1.1	20 10		Dust, Sea Salt	in tooldy	01710	2011 proboint	1 011111010 01 01., 201	
MRC			1 x 1			BB smoke, Dust,			2003-present	this work	
	,	*				Sea Salt, ABF	-	-	2000-present	UNS WOLK	

# Table 2. Parameters representing microphysical and optical properties of aerosol species from the four aerosol reanalyses.

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	Microphysics (sectional size	bins in radius o	r bulk effectiv	ve radius in	Optical parameters at 550nm for the corresponding size bins (single scattering albedo, mass extinction efficiency m2/g , and shape for dry particle)						
Species Models	Dust	Sea salt	sulfate/ABF	BB smoke /OC/OM	вс	Dust	Sea salt	sulfate/ ABF	BB smoke /OC/OM	вс	
CAMSRA	0.55 - 0.9,	0.03- 0.5, 0.5 -5, 5 - 20	0.005 - 20	OM: 0.005 - 20	0.005 - 0.5	0.90; 0.92	1.0; 0.73 1.0; 0.14 1.0; 0.04 sphere	1.0; 4.33	OM: 0.89; 2.76 sphere	0.21; 9.4 <sup>.</sup> sphere	
MERRA-2	1.0 - 1.8, 1.8 - 3.0, 3.0 - 6.0,	0.03 - 0.1, 0.1 - 0.5, 0.5 - 1.5, 1.5 - 5.0, 5.0 - 10	Bulk, 0.16	OC: Bulk 0.09	Bulk, 0.04	0.92; 0.64 0.89; 0.33 0.83; 0.17 0.77; 0.08	1.0; 0.73 1.0; 3.48 1.0; 0.74 1.0; 0.30 1.0; 0.10 sphere		OC: 0.96; 2.67 sphere	0.21; 9.28 sphere	
NAAPS-RAv1	Bulk, 2.5	Bulk, 1.5	Bulk, 0.14	Smoke: Bulk, 0.17	N/A		0.99; 1.42 sphere	ABF 0.9; 3.48 sphere	Smoke: 0.89; 4.48 sphere	N/A	
JRAero	1.00 – 1.59, 1.59 – 2.51, 2.51 – 3.98, 3.98 – 6.30,	0.159 – 0.251, 0.251 – 0.398,	Bulk, 0.15	OC: Bulk, 0.18	Bulk, 0.18	0.98; 3.36 0.97; 3.32 0.94; 1.45 0.90; 0.82 0.86; 0.48 0.81; 0.29 0.75; 0.18 0.68; 0.11	1.0; 0.17 1.0; 0.56 1.0; 1.36 1.0; 1.97 1.0; 1.53 1.0; 0.54 1.0; 0.39 1.0; 0.23 1.0; 0.14 1.0; 0.08 sphere		0.96; 1.60 sphere	0.16; 5.3 sphere	

 265 Table 3. Hygroscopic enhancement factor (*f*) at different relative humidity (RH) levels for

266 various aerosol species in the four RAs. In MERRA-2, f for sea salt varies with size bins, thus a

**267** range for *f* is presented here. Notably, NAAPS-RA v1 does not explicitly contain BC species.

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268 <u>More specific details can be found in the references provided in Table 1.</u>

RH (%)		Sea	salt		Sulfate/ABF				BB smoke/OM/OC				BC			
	CAMSRA	MERRA2	NAAPSRA	JRAero	CAMSRA	MERRA2	NAAPSRA	JRAero	CAMSRA	MERRA2	NAAPSRA	JRAero	CAMSRA	MERRA2	JRAero	
<30	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	
30	1.00	1.17-1.22	1.00	1.36	1.00	1.23	1.00	1.24	1.00	1.14	1.00	1.12	1.00	1.00	1.00	
40	1.44	1.21-1.28	1.07	1.48	1.17	1.31	1.08	1.32	1.17	1.19	1.03	1.16	1.00	1.00	1.00	
50	1.56	1.26-1.35	1.17	1.60	1.22	1.39	1.18	1.40	1.20	1.24	1.06	1.20	1.00	1.00	1.00	
60	1.67	1.33-1.44	1.29	1.70	1.28	1.46	1.32	1.45	1.30	1.29	1.11	1.30	1.00	1.01	1.00	
70	1.80	1.44-1.56	1.48	1.80	1.36	1.54	1.53	1.50	1.40	1.34	1.16	1.40	1.00	1.03	1.00	
80	1.99	1.60-1.77	1.78	2.00	1.49	1.64	1.87	1.60	1.50	1.44	1.25	1.50	1.20	1.19	1.20	
85	2.13	1.74-1.93	2.03	2.20	1.58	1.69	2.16	1.70	1.55	1.52	1.32	1.55	1.30	1.30	1.30	
90	2.36	1.96-2.19	2.45	2.40	1.73	1.77	2.65	1.80	1.60	1.64	1.42	1.60	1.40	1.41	1.40	
95	2.88	2.43-2.74	3.37	2.90	2.09	1.91	3.74	1.90	1.80	1.88	1.61	1.80	1.50	1.54	1.50	

# 270 2.3 AERONET

AERONET is a global ground-based sun photometer network managed by NASA. Sun and sky radiance at multiple wavelengths, covering the near-ultraviolet to near-infrared, are measured (Holben et al., 1998). Version 3 Level 2 AERONET daily data (Giles et al., 2019), which are cloud-screened and quality-assured, are used in this study. The estimated uncertainty in AERONET measured AOD, due primarily to calibration uncertainty, is ~0.01-0.02 at optical airmass of one for network field instruments (with the highest errors in the UV; Eck et al., 1999).

277 The 550 nm FM and CM AODs and total AODs are derived with the Spectral Deconvolution Method (SDA; O'Neill et al. 2003). The AERONET SDA product has been verified using in situ 278 279 measurements (see for example Kaku et al., 2014). The spectral separation of FM and CM particles is determined based on their distinctive optical properties and complete size distributions. As part 280 of this separation, a diameter of approximately 1µm serves as an approximate threshold to 281 differentiate FM and CM particles. This optical separation is different from the sub-micron fraction 282 (SMF) method that uses a specified cutoff radius of the particle size distribution in the AERONET 283 284 (AOD & sky radiance) inversion and allows more data to be available compared to the SMF 285 method. The FM fraction based on SDA is generally comparable and slightly greater than SMF 286 (O'Neill et al., 2023).

This study uses AERONET sites that have more than 5 years of observations and more than 1000 daily data between 2011 and 2019 for verification purposes. Monthly AOD was derived for months that have more than 15 days of daily data. Then only sites with more than 45 total number of months (upper three quartiles of sites regarding total number of monthly data) were selected. This resulted in a total number of 200 sites globally. AThe list of sites along with latitude/longitude coordinates and elevation details for the site namesstudied regions is availableaccessible in Table S1-and. Additionally, the locations of theseall sites can be foundidentified in Figure 8.

294 2.4 MODIS AOD

295 MODIS AOD, used for global AOD distribution evaluation of the RAs, was based on Collection 296 6.1 Dark Target and Deep Blue retrieval products (Levy et al., 2013). Additional quality 297 control Three MODIS AOD products are used as reference datasets to show global distribution of 298 AOD climatology and the divergence among the retrieval products in comparison with the RAs. 299 The level 3 MODIS AOD data for Dark Target (DT) were constructed using collection 6.1 Aqua 300 MODIS level 2 DT data. The level 2 DT MODIS aerosol retrievals are available at a 10×10 km<sup>2</sup> 301 spatial resolution over both land and ocean. These aerosol retrievals were initially averaged on a daily basis at a spatial resolution of  $0.5 \times 0.5^{\circ}$  lat/lon. Only data with a quality flag of "marginal" 302 303 or better were used in the analysis. Additionally, retrievals with a cloud fraction larger than 80% 304 were excluded to minimize cloud contamination, as suggested by Zhang et al. (2005). The level 3 305 DT MODIS AOD data  $(0.5 \times 0.5^{\circ} \text{ lat/lon})$  were then constructed using the daily averaged AOD 306 data. 307 Similar approaches were applied to C6.1 Aqua MODIS level 2 Deep Blue (DB) AOD data. Unlike the MODIS DT aerosol retrievals, which are available over regions with low surface 308 309 reflectance, the DB retrievals are also available over some bright regions, such as desert regions. 310 No over-ocean aerosol retrievals, however, are included in the MODIS level 2 aerosol data. The 311 level 2 DB MODIS aerosol data were used to construct daily averages at a spatial resolution of 312  $0.5 \times 0.5^{\circ}$  (lat/lon). No quality flag and cloud fraction thresholds were applied. The level 3 DB MODIS AOD data (0.5×0.5° lat/lon) were constructed using the daily averaged AOD data. 313 314 The third MODIS AOD product is a data-assimilation-quality AOD dataset. It was based on C6.1 315 DT and DB retrieval products (Levy et al., 2013). Strict quality control and bias-correction processes were applied as described in Zhang and Reid (2006) and Shi et al. (2011) for over water, 316 317 Hyer et al. (2011) for over land, and Shi et al. (2013) for over desert regions. These quality control 318 processes were updated for the Collection 6C6.1 data and the final -MODIS C6.1 AOD (550 nm) 319 data is a level 3 product with 1°x1° latitude/longitudelat/lon spatial and 6-hourly temporal 320 resolution. Those This product has a cut-off at 40°S to filter out potential cloud-contaminated data 321 south of this latitude. The 6-hour-averaged MODIS-AOD data were then binned into monthly 322 means. Note that MODIS AOD products are well known to low bias significant aerosol events 323 (Reid et al., 2022; Gumber et al., 2023), which could result in a slightly low AOD climatology, 324 especially in source regions.

Note that MODIS AOD products are well known to low bias significant aerosol events (e.g., Reid
 et al., 2022; Gumber et al., 2023) and slightly high bias clean environment (e.g. Wei et al., 2019),
 which could affect AOD climatology to some degree.

328 2.5 Analysis Method

This study aims to investigate the <u>diversitydivergence</u> and utility of RAs for climate-scale studies by exploring the AOD at 550 nm. To achieve this goal, the AOD data from the RAs, as well as MODIS, were spatially and temporally binned into 1°x1° degrees and monthly resolutions. For the purpose of verification and intercomparison analysis, only the data between 2011 and 2019 were used as that is the <u>time</u>-period when all the RAs have data. The study focuses on the 550 nm AOD parameter since it is available for all the-four aerosol RAs and MODIS. Furthermore, the
 AERONET FM and CM AODs at 550 nm were obtained using the SDA method described in Sect.
 2.3.

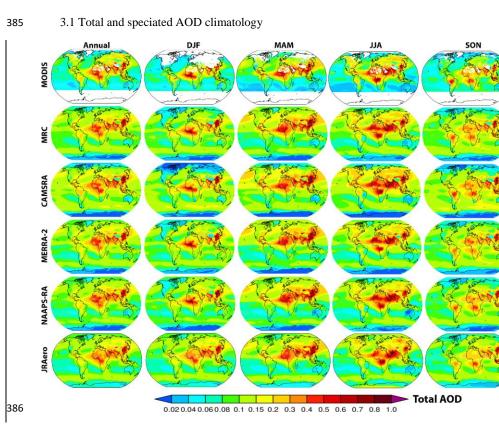
The study examines the performance of RAs globally and regionally. Sixteen regions, including 337 the globe, are defined for regional aerosol property analysis. They include East Asia, Southeast 338 Asia, South Asia, Maritime Continent, Australia, Southwest Asia, Europe, Northwest Africa, 339 340 South Africa, West North America, East North America, Central America, South America, as 341 indicated by the rectangular boxes in Figure 5, and Arctic (north of 70°N), and Antarctic (south of 342 75°S). There is no AERONET site satisfying site selection criteria as described in Section 2.3 in 343 the Arctic and Antarctic, so these two regions are not included excluded for regional verification though they are included in other analyses. 344

345 Regarding the aerosol species, the study focuses on BB smoke, ABF in NAAPS-RA, and its 346 equivalent of sulfate for MERRA-2, CAMSRA, and JRAero, as well as dust and sea salt. The 347 definition of species follows the ICAP practices (Sessions et al., 2015; Xian et al., 2019) for the 348 operational counterparts of these RAs and previous applications of these RAs (e.g., Xian et al., 349 2022), in which the sum of Organic Matter (OM) and BC AODs from CAMSRA, and the sum of 350 OC and BC AODs from MERRA-2 and JRAero, is used to approximate BB smoke AODs. 351 Although this separation of species may be somewhat arbitrary, the study takes into account the fact that different aerosol types and sources may be represented differently in each RA. For 352 353 example, the NAAPS-RA model characterizes aerosol species by emission source rather than chemical speciation, which makes it unique. In contrast, CAMSRA, MERRA-2, and JRAero 354 characterize OM or OC, BC, and inorganic species, merging contributions from various 355 356 anthropogenic, biomass burning and biogenic sources.

357 The study also assumes that all sea salt and dust are CM, while other aerosol species are FM. The segregation of sea salt and dust to the CM category is based on the fact that only a small portion 358 of -total sea salt or dust AOD at 550nm are attributed to their FM components. For example, FM 359 sea salt represents about 17%, 10% and 11% of total sea salt AOD globally in MERRA-2, 360 CAMSRA and JRAero respectively. The numbers are about 30%, 39% and 32% for dust. While 361 362 FM fraction of dust during dust storms in Africa varies between 20-25% according to AERONET. 363 The FM fraction of dust from MERRA-2, CAMSRA and JRAero might be biased high as these 364 global models tend to overestimate FM dust and underestimate CM dust (for example O'Sullivan 365 et al., 2020; Kramer et al., 2020). In contrast, NAAPS-RA assumes all sea salt and dust are CM. 366 Verification results based on the FM and CM AODs derived using the FM fractions of sea salt and 367 dust from MERRA-2, CAMSRA and JRAero can be found in the supplemental material- (Fig. S2-368 4). Generally, the validation of FM and CM AODs with AERONET data shows a degradation in 369 performance for the three RAs compared to the verification results presented below, as discussed 370 in section 3.3.1.

371 AOD validation results for total, FM, and CM AOD regarding bias, root mean square error 372 (RMSE), and coefficient of determination ( $r^2$ ) for monthly mean AODs are presented. 373 For every AERONET site, the time series of monthly modal AOD from each RA is first extracted 374 from the model grid that encompasses the site's location. Bias, root mean square error (RMSE), 375 and coefficient of determination  $(r^2)$  are then computed for each site and each RA. The regional 376 validation outcome is derived from the average of validation statistics across all sites within the 377 region (see Table S1 for the sites included in each region). Following the criteria for site selection 378 outlined in section 2.3, only 200 sites are available globally, and certain regions have only a few 379 sites (a minimum of three sites, such as in South Africa) to represent the entire region; hence, no 380 site weighting within a region is applied. It is acknowledged that this averaging method could bias 381 the global validation result toward regions densely populated with sites, notably North America 382 and Europe. The AOD validation results for total, FM, and CM AOD at 550nm are presented 383 accordingly.

#### 384 3. Results



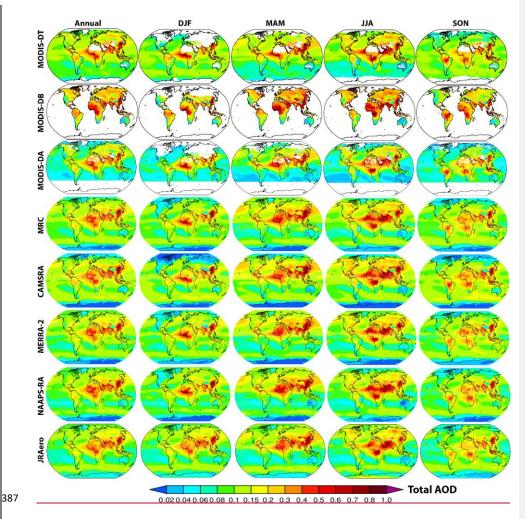
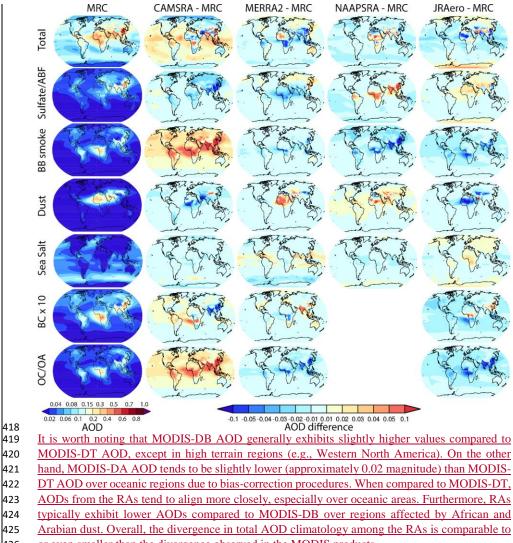


Figure 1. Annual and seasonal total 550nm AOD climatology from three MODIS\_products, the
four RAs, and the MRC over 2003-2019, except JRAero for 2011-2019. The white area-MODISDA is the data-assimilation-quality AOD dataset described in Section 2.4. In the MODIS plots-,
the white area means a lack of data-attributed to either none valid-retrievals or quality-control
filtering. Notably, MODIS-DB data is only available over land.

The climatological annual and seasonal mean total AODs at 550nm from <u>the three MODIS AOD</u> datasets and the four aerosol RAs and <u>their consensus (the MRC)</u> are presented in Figure 1. In general, there are very similar spatial AOD distribution patterns and AOD magnitude among the 396 RAs and MODIS datasets for all four seasons. This is expected as MODIS total AOD is assimilated 397 into all of these RA products as well as used to tune the model components such as emissions. High AOD regions include the dust-dominated Sahara in Mar-Apr-May (MAM) and Jun-Jul-Aug 398 (JJA), Sahel in Dec-Jan-Feb (DJF) and MAM, Southwest Asia and Taklamakan in MAM and JJA, 399 anthropogenic pollution-dominated East Asia and South Asia throughout the year, BB smoke-400 401 dominated South Africa, South America in JJA and Sep-Oct-Nov (SON), Southeast Asia in MAM, Maritime Continent in SON, and high-latitude North America and Eurasia in JJA. For the annual 402 403 mean, MODIS AOD is AODs from all the three products are relatively high compared to the MRC 404 in the northern hemisphere's high latitudes due to seasonal sampling bias. MODIS was able to 405 retrieve AOD during biomass burning active season, i.e. boreal Summer-to-Fall, but it couldn't 406 retrieve AOD during northern winter in the high latitudes due to the lack of sunlight- and the high 407 snow/ice coverage. The high AOD over high-latitude Eurasia and North America in MODIS 408 annual mean is a general reflection of MODIS summertime AOD, which is captured by all the 409 RAs in their summertime mean AODs. It is also noted that all the RAs have slightly higher AOD 410 (on the order of 0.02) over the ocean than MODIS QAed product here. MODIS AOD products are 411 well known to low bias significant aerosol events (e.g., Reid et al., 2022; Gumber et al., 2023), 412 which could lower the mean state of AOD. The slightly lower MODIS AOD compared to the RAs 413 could also be related to clear-sky and contextual bias (Zhang et al., 2009), as MODIS AOD 414 retrieval is only available under clear sky conditions, while all the RAs include all sky conditions. 415 Sea salt and dust emissions are often associated with cloudy synoptic weather systems, and 416 hygroscopic aerosol species, such as sulfate, sea salt, and BB smoke, can potentially grow larger 417 in size in a moister environment, introducing a higher all-sky AOD than the clear sky AOD.



426 <u>or even smaller than the divergence observed in the MODIS products.</u>

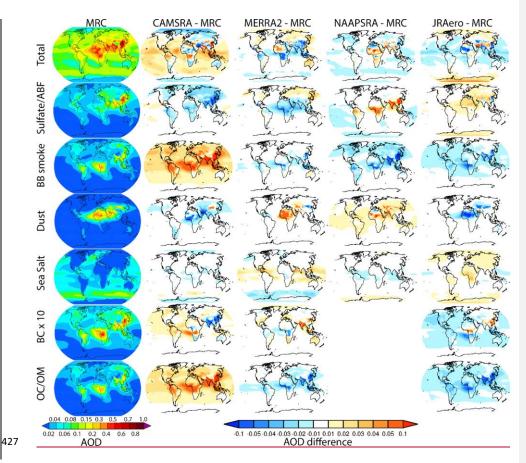


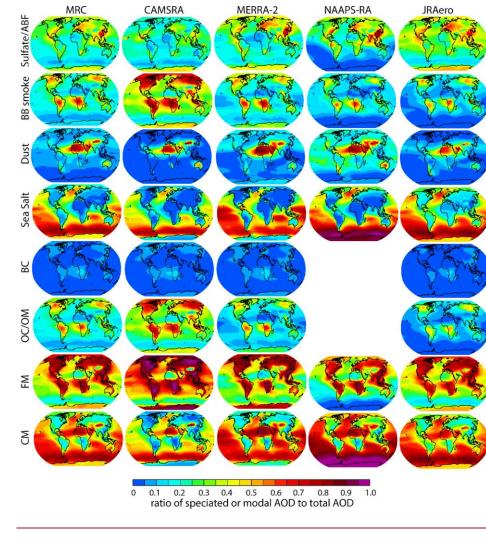
Figure 2. Annual mean total and speciated AODs of the MRC and the AOD difference between
the individual RA and the MRC based on the 2011-2019 average. <u>BB smoke is approximated as</u>
the sum of OC/OM and BC in CAMSRA, MERRA-2 and JRAero.

Previous experience with multi-model ensembles suggests that the consensus of multi-models, in 431 432 general, shows better skill than individual contributing models (Sessions et al., 2015; Xian et al., 2019; Reid et al., 2022). Similar verification conclusion is also drawn in Section 3.3. Therefore, 433 434 the total and speciated AODs from the MRC based on the 2011-2019 average are used as a baseline 435 here and are shown in Figure 2. As expected, sulfate/ABF AOD is relatively high over populationdense and industrially polluted regions, dust AOD is high over major desert and arid regions, and 436 sea salt AOD is relatively high over mid-to-high latitude oceans. BB smoke and its components 437 438 BC and OC/OAOM are relatively high over South Africa, South America, Southeast Asia, the 439 Maritime continent, and Siberia, North American high latitudes major BB source regions. BC and 440 OC/OAOM AOD are also relatively high over South Asia and East Asia, where sources other than BB, such as anthropogenic emission, are the main contributors, as suggested by contrasting smoke
AOD contribution to the total AOD between NAAPS-RA and other RAs in these regions
(FiguresFigs 3 and 10. Noting that smoke AOD is driven by BB in NAAPS-RA, while smoke
AOD is a sum of BC and OC/OAOM from the other RAs).

445 Shown also in Figure 2 are the total and speciated AOD differences between the individual RA and the MRC. For total AOD, CAMSRA is apparently higher than the other three RAs over the 446 447 ocean, which is consistent with the findings on its operational counterpart of high biased FM AOD verified with Maritime Aerosol Network over the ocean in Reid et al. (2022). This high bias is 448 449 attributed to its universally higher OAOM/smoke AOD compared to other RAs, and suggests that 450 CAMSRA may have higher BB emissions and/or less efficient removals higher secondary 451 production of OM compared to the other RAs. Sulfate AOD is relatively low in CAMSRA except 452 for some highly biased hotspots around outgassing volcanoes (in particular Mauna Loa and near 453 Mexico City) as mentioned in Inness et al (2019). Differences in species definitions affect the 454 comparison with NAAPS-RA: NAAPS-RA ABF AOD is higher than sulfate AOD in other RAs 455 especially in East Asia, South Asia, central Africa, and north South America, and these deviations 456 are counterbalanced by opposite deviations in the BB AOD. This is expected as ABF in NAAPS-457 RA includes biogenic and anthropogenic primary and secondary aerosolsadditional aerosol 458 sources besides sulfate-, and some of these sources are included in the BB AOD for other models. 459 For dust AOD, MERRA-2 is relatively higher over north Africa and the Arabian Peninsula and NAAPS-RA is relatively higher over most regions, including oceanic areas, while CAMSRA and 460 461 JRAero are relatively lower over most regions except around Gobi desert for CAMSRA and Iran 462 for JRAero. As for sea salt AOD, MERRA-2 is relatively higher over the tropical oceans, and 463 lower over the southern ocean. JRAero sea salt AOD is relatively higher over most continents, which is probably unphysical. 464

The differences in speciated AOD result in significant variations in their contributions to the total 465 466 AOD, as illustrated in Figure 3. For instance, the considerably higher BB smoke AOD in CAMSRA compared to other RAs makes BB smoke the predominant contributor to total AOD in 467 the CAMSRA over most continents, adjacent water bodies, and polar regions, except for regions 468 469 where dust is dominant. Sulfate AOD, on the other hand, contributes more to the total AOD, 470 particularly over oceanic regions in the JRAero compared to other RAs. Both MERRA-2 and 471 JRAero exhibit higher sulfate contributions along the western coasts of South America and North 472 America, suggesting possible increased production of dimethyl sulfide (DMS) in those areas. Dust 473 AOD, on the other hand, contributes more to the total AOD particularly over oceanic regions in 474 NAAPS-RA compared to the other RAs. Sea salt AOD is found to contribute more to the total 475 AOD in the high-latitude oceans and the Antarctic in NAAPS-RA compared to the other RAs. The 476 OC/OAOM AOD contribution to the total AOD closely mirrors the distribution of BB smoke, as 477 anticipated. The contribution of BC to the total AOD is generally small, ranging between 5-10% 478 in BB regions, except for central South Africa where it reaches 10-15%. Despite the higher ratio 479 of BB smoke AOD to total AOD ratio-in CAMSRA, the ratio of BC to total AOD over East Asia 480 and South Asia is smaller in CAMSRA compared to MERRA-2 and JRAero, suggesting that BC 481 emissions from anthropogenic sources maybe lower in CAMSRA- (also Fig. 2). Finally, the

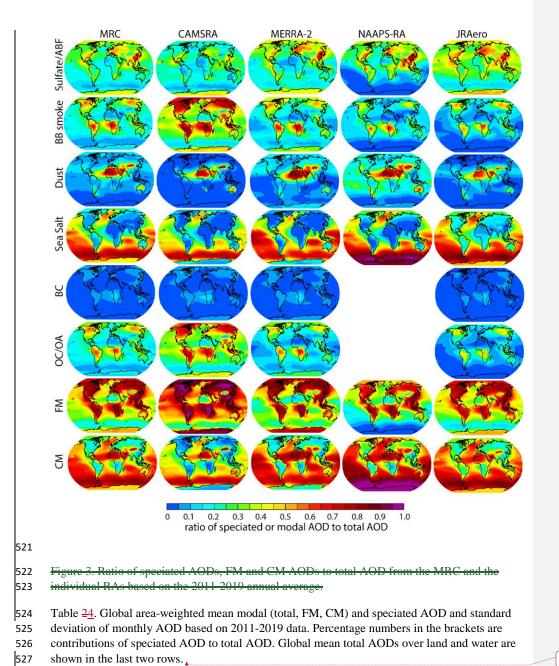
contributions of FM and CM AOD to the total AOD are also depicted in Figure 3. It is consistent 482 483 among the RAs that FM is the dominant contributor over most land regions except for regions where dust is dominant, such as North Africa, the Arabian Peninsula, the Middle East, and the 484 Gobi. In all the RAs, CM is the dominant contributor over oceanic regions, except for regions 485 486 influenced by continental BB smoke and pollution outflow. The contribution of CM in CAMSRA is generally smaller in tropical to mid-latitude oceans compared to other RAs, due to its higher 487 contribution from BB smoke. It is also noted that CM is dominant over FM in the Antarctic in 488 NAAPS-RA, while FM is dominant in the Antarctic in the other three RAs, though total AOD is 489 490 very small (annual and seasonal means < 0.04 from MRC) and hard to validate due to lack of observational data. 491



493 Figure 3. Ratio of speciated AODs, FM and CM AODs to total AOD from the MRC and the
 494 individual RAs based on the 2011-2019 annual average.

Table 24 provides a summary of global-average total AOD and speciated AODs, as well as the contributions of speciated AOD to total AOD for all the RAs. Overall, the annual and global mean total AODs are similar, hovering around 0.14 for most RAs. However, CAMSRA stands out with a slightly higher total AOD of 0.151, which compared to the MRC is 0.012 higher, while the differences between the other RAs and MRC are within ±0.005. Total AODs over land show minimal variation among the RAs, likely due to the cancellation of high and low biased speciated
 AODs. Over water, CAMSRA exhibits slightly higher AOD compared to other RAsAll land and
 ocean mean AODs are within 0.006 of the MRC with the exception of CAMSRA over ocean,
 which is higher than the MRC by +0.024.

504 Speciated AODs, especially smoke AOD and OAOM/OC AOD display greater 505 diversitydivergence among the RAs. Smoke and OAOM AODs from CAMSRA are 2-3 times 506 higher than those from the other RAs. Smoke AOD contributes to 41% of total AOD in CAMSRA, 507 while ranging from 16%-22% in other RAs. Moreover, the standard deviation of smoke and 508 OAOM AODs with respect to the 12 months is also higher in CAMSRA than in other RAs. The 509 contribution of dust AOD to total AOD varies from 13% to 28% for all the RAs, with NAAPS 510 dust AOD being the highest among the RAs and about 2 times that of CAMSRA, which has the 511 lowest dust AOD among the RAs. The contribution of sulfate/ABF AOD to total AOD ranges 512 from 23% to 34%, with the highest contribution observed in JRAero-, even larger than the ABF 513 AOD contribution in NAAPS-RA. Sea salt AOD contributes 25% to 35% to total AOD in the RAs 514 with JRAero being the highest. BC AOD, on the other hand, contributes only 3% to 4% of total 515 AOD across the RAs. The FM's contribution to the overall AOD varies across different datasets. 516 In MERRA-2, NAAPS-RA, and JRAero, FM accounts for 44% to 51% of the total AOD. However, in CAMSRA, its contribution is notably higher at 63%, primarily due to its significant 517 518 contribution from **BBOM**. Conversely, CM's contribution to total AOD is consistent across the three RAs, ranging from 49% to 56%. In contrast, CM's contribution is lower, at 37%, in 519 520 CAMSRA.



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		glo	obal mean A	OD		AOD standard deviation w.r.t. 12 months						
	CAMSRA	MERRA2	NAAPSRA	JRAero	MRC	CAMSRA	MERRA2	NAAPSRA	JRAero	MRC		
total	0.151	0.137	0.134	0.134	0.139	0.018	0.010	0.011	0.012	0.013		
dust	0.019 (13%)	0.029 (21%)	0.037 (28%)	0.021 (16%)	0.026 (19%)	0.008	0.009	0.009	0.009	0.008		
sea salt	0.037 (25%)	0.041 (30%)	0.038 (28%)	0.045 (34%)	0.040 (29%)	0.001	0.001	0.003	0.002	0.001		
sulfate/ABF	0.034 (23%)	0.037 (27%)	0.037 (28%)	0.046 (34%)	0.039 (28%)	0.002	0.001	0.001	0.002	0.001		
smoke	0.062 (41%)	0.030 (22%)	0.022 (16%)	0.022 (16%)	0.034 (24%)	0.009	0.007	0.007	0.007	0.007		
BC x 10	0.061 (4%)	0.059 (4%)	-	0.044 (3%)	0.054 (4%)	0.013	0.009	-	0.008	0.009		
OC/OA	0.056 (37%)	0.024 (18%)	-	0.018 (13%)	0.033 (24%)	0.007	0.006	-	0.006	0.006		
FM	0.096 (63%)	0.067 (49%)	0.059 (44%)	0.068 (51%)	0.073 (53%)							
СМ	0.056 (37%)	0.070 (51%)	0.075 (56%)	0.066 (49%)	0.066 (47%)							
land total	0.180	0.174	0.175	0.176	0.176							
water total	0.136	0.118	0.112	0.111	0.112							

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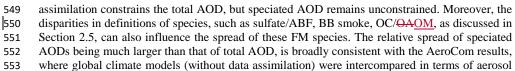
		glo	obal mean A	DD		AOD standard deviation w.r.t. 12 months						
	CAMSRA	MERRA2	NAAPSRA	JRAero	MRC	CAMSRA	MERRA2	NAAPSRA	JRAero	MRC		
total	0.151	0.137	0.134	0.134	0.139	0.018	0.010	0.011	0.012	0.01		
dust	0.019 (13%)	0.029 (21%)	0.037 (28%)	0.021 (16%)	0.026 (19%)	0.008	0.009	0.009	0.009	0.00		
sea salt	0.037 (25%)	0.041 (30%)	0.038 (28%)	0.045 (34%)	0.040 (29%)	0.001	0.001	0.003	0.002	0.00		
sulfate/ABF	0.034 (23%)	0.037 (27%)	0.037 (28%)	0.046 (34%)	0.039 (28%)	0.002	0.001	0.001	0.002	0.00		
smoke	0.062 (41%)	0.030 (22%)	0.022 (16%)	0.022 (16%)	0.034 (24%)	0.009	0.007	0.007	0.007	0.00		
BC x 10	0.061 (4%)	0.059 (4%)	-	0.044 (3%)	0.054 (4%)	0.013	0.009	-	0.008	0.00		
OC/OM	0.056 (37%)	0.024 (18%)	-	0.018 (13%)	0.033 (24%)	0.007	0.006	-	0.006	0.00		
FM	0.096 (63%)	0.067 (49%)	0.059 (44%)	0.068 (51%)	0.073 (53%)							
CM	0.056 (37%)	0.070 (51%)	0.075 (56%)	0.066 (49%)	0.066 (47%)							
land total	0.180	0.174	0.175	0.176	0.176							
water total	0.136	0.118	0.112	0.111	0.112							

# 530 **3.2** Geographical diversity divergence of speciated AOD among the four reanalyses RAs

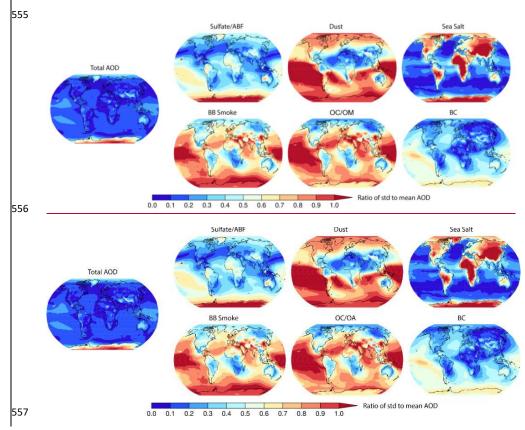
The diversitydivergence of the global-average total and speciated AODs is already documented in 531 532 Table 24. Figure 4 provides the geographical distribution of the relative spread of speciated annual mean AODs from the RAs to their means. Spread, in this context, is defined as the ratio of the 533 534 standard deviation of the RAs AODs to their mean. It is noteworthy that the relative spread of total AOD from the four RAs is generally small, except for polar regions and specific hotspots where 535 known issues exist. For instance, biases in CAMSRA AOD have been identified over Hawaii and 536 Mexico's volcanic outgassing regions. In polar regions, there are limited satellite observations to 537 constrain model fields, resulting in a larger spread, which is consistent with the findings of Xian 538 539 et al. (2022) on AODs from CAMSRA, MERRA-2 and NAAPS-RA over the Arctic. Similarly, 540 over high terrains with snow and ice covers, such as the Himalayas and the Andes, and over desert regions, such as the Australian deserts, and the Bodele Depression region in the Sahara, both 541 retrievals and models face challenges, leading to a larger spread. Moreover, over the Maritime 542 Continent, where high cloud coverage poses challenges to remote sensing retrievals for both AOD 543 544 and BB smoke emissions, the spread is also relatively large.

545 The aforementioned characteristics are also evident in the spread of speciated AODs. However, 546 the spreads of the speciated AODs among the RAs are much larger compared to the total AOD, 547 particularly in regions that are remote from aerosol sources. This suggests that the efficiency of

removal processes during long-range transport may differ. This is also relevant to the fact that data



554 optical properties and life cycles (Kinne et al., 2006; Textor et al., 2006; Gliß et al., 2021).



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Figure 4. Spread of total and speciated climatological annual-mean AOD among the four RAs.Spread here is defined as the ratio of the standard deviation of the RA AODs to their mean.

### 560 **3.3. Evaluation with AERONET AOD**

This section presents evaluation of the monthly performance of the four RAs plus the
 consensus<u>MRC</u> at the AERONET sites on regional and global scales. Both skill and consistency
 of the different RAs and consensus are evaluated.

564 3.3.1 Bias, RMSE, and correlation between the RAs and AERONET

566 The regional and global mean modal AOD bias, RMSE, and squarecoefficient of the 567 correlationsdetermination for the four RAs and the MRC are shown as bar graphs on global maps 568 in Figures 5, 6 and 7. Regarding regional bias, all the RAs except for CAMSRA, have large negative biases (on the order of -0.1) in total AOD over Southeast Asia, South Asia, and the 569 Maritime continent (Figure 5). The much smaller negative bias in total AOD over these regions in 570 CAMSRA is a result of the cancelation of a positive bias in FM, possibly due to high biased 571 OAOM/smoke AOD, and a negative bias in CM. The large negative biases over these regions in 572 the other RAs are mainly attributed to large negative biases in FM AOD in general. It is also noted 573 that CAMSRA is biased relatively high in total AOD due to high FM bias over East Asia. Over 574 other regions and the globe, all the RAs have relatively small biases and in general slight positive 575 biases, with CAMSRA having the largest positive bias, due mainly to relatively high 576 577 OAOM/smoke AOD. The cancellation effect of positive FM bias and negative CM bias in CAMSRA are also visible. 578

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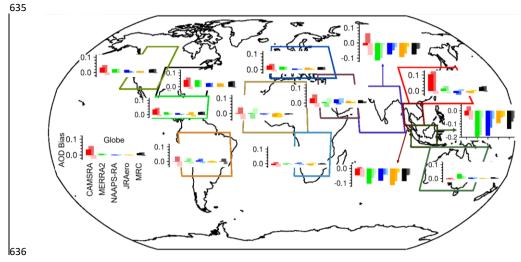
580 Total AOD RMSEs are relatively high over all Asian regions and North Africa compared to other 581 regions for all the RAs (Figure 6). The contribution of FM to total AOD RMSE is larger than that 582 from CM globally, except over dust-influenced regionsregion, including North Africa and, for most models, Southwest Asia and Central America. The correlations of total AOD between the 583 RAs and AERONET data are mostly reasonable for all the regions (Figure 7). Some relatively 584 low-performance regions (total AOD  $r^2$  less than 0.60 for at least one RA) include South Asia, 585 Southwest Asia, Australia, Europe, and East Asia. The relatively low correlations over Australia 586 587 and Europe are due to the low climatological mean and variance. While the other low-performance 588 regions are all mixed pollution and dust environment that is challenging for all RAs. Some relatively high-performance regions (total AOD  $r^2$  greater than 0.9085 for at least two RA 589 590 members) include Central America, Peninsula Southeast Asia, and Maritime Continent. Total and 591 CM AOD  $r^2$  are high over Central America, because it is a receptor region for African dust, and 592 RAs perform well in general during long-range transport over ocean where data assimilation is very effective in correcting model AOD fields. Total and FM AOD r<sup>2</sup> are high over Peninsula 593 Southeast Asia, and Maritime Continent, because suggesting the RAs can capture the large 594 interannual variabilities of the regional dominant aerosol species, BB smoke, have large 595 596 interannual variabilities, due to associated with the impact of ENSO on fire activities in the regions (e.g., Reid et al., 2012; Xian, et al., 2013). Overall, the MRC exhibits superior r<sup>2</sup> compared to 597 individual RAs for modal AODs regionally and globally. 598 599

For remote marine sites, including Ascension Island in the mid-basin of south Atlantic, Ragged 600 601 Point in the western Tropical Atlantic, Mauna Loa in Hawaii, MCO-Hanimaadhoo in the north Indian Ocean, and REUNION\_DENIS in the south Indian Ocean, the RAs exhibit similar 602 performance at these sites as they do over the upwind land or coastal regions (Fig. S1). An 603 exception is MauraMauna Loa. - MauraMauna Loa is situated at an elevation of 3.4 km, well above 604 the marine boundary layer and remote from continental sources. At this location, all the RAs 605 606 exhibit a significant positive bias. One possible explanation for this bias is the topographic effect, 607 as the coarse spatial resolutions of the models may not be able to resolve the site's high elevation-608 or its sharp elevation gradient compared to the surroundings. Additionally, uncertainties in the 609 removal processes during long-range transport may also be contributing to the high bias. It is also 610 worth noting that all the RAs do especially well at the Ragged Point site, with total AOD  $r^2$  close

to or higher than 0.92. This site is a receptor site of African dust in the Western Tropical Atlantic.
 This suggests that the RAs capture the long range-transport of dust from Africa quite well. This is
 related to the fact that data assimilation systems have more chance to correct the model fields with

- 614 <u>observations in the long-range transport over the ocean.</u>
- 615

When considering the contribution of dust and sea salt aerosols to FM AOD in CAMSRA, 616 MERRA-2 and JRAero, the verification statistics (bias, RMSE and  $r^2$ ) for the total AOD of these 617 618 RAs remain unchanged as expected (Fig. S2, S3, S4). However, there is a noticeable shift in the positive bias of FM AOD (and negative bias of CM AOD) for these RAs, particularly in regions 619 620 influenced by dust, such as North Africa, the Arabian Peninsula, East Asia, Central America, South Asia, and Europe. Specifically, the positive bias in FM AOD becomes more pronounced, and the 621 negative bias in CM AOD becomes more negative in these regions, especially for CAMSRA. It's 622 623 worth noting that in MERRA-2, there is a change in sign, where the FM AOD bias switches from negative to positive in North Africa and the Arabian Peninsula, while the CM AOD bias changes 624 from positive to negative in these regions. Additionally, the negative FM AOD bias becomes 625 smaller, however the negative CM AOD bias worsens in South Asia within both MERRA-2 and 626 627 JRAero datasets (Fig. S2). In general, when taking into account the contribution of dust and sea salt aerosols to FM AOD (by default, dust and sea salt AODs are treated as CM AODs in this 628 629 study) in CAMSRA, MERRA-2, and JRAero, we observe a worsening of both FM and CM AOD 630 biases in these three datasets. Similarly, the RMSE for both FM and CM AODs over regions influenced by dust deteriorates as well (Fig. S3). The  $r^2$  for FM and CM AODs in these regions 631 also worsens overall, with the exception of an improvement in FM AOD over Central America. 632 633 FM sea salt's impact on the verification score is small as the majority of AERONET sites are on land and FM sea salt only contributes on the order of  $\sim 10\%$  to total sea salt AOD in the three RAs. 634



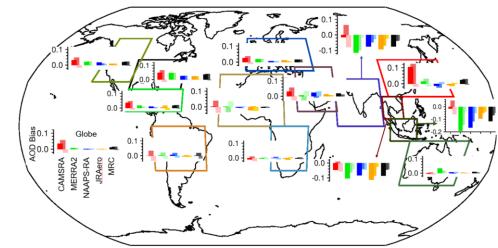
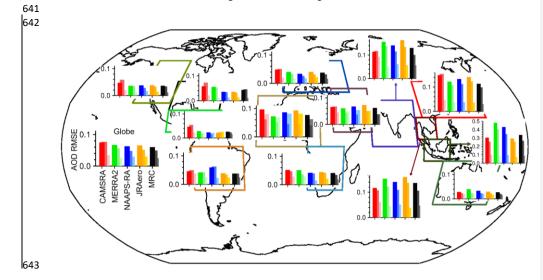
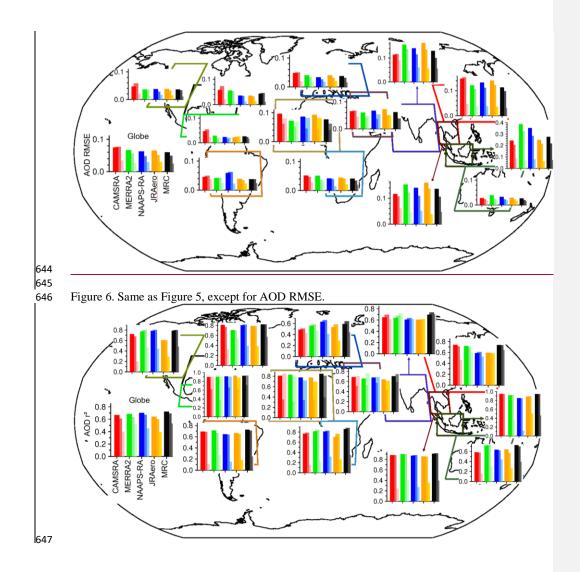
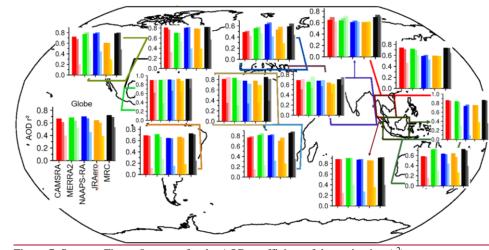




Figure 5. Regional total, FM, and CM AOD biases for the four reanalyses and the MRC compared with AERONET data. Each grouped bars in the same color system present total, FM, and CM AOD biases from left to right (also dark to light).







651

Figure 7. Same as Figure 5, except for the AOD coefficient of determination  $(r^2)$ .

652 3.3.2 Rankings of the RAs with respect to validation statistics

653 To expand the validation result from regional averages to individual sites, including remote sites that are not included in the regional analysis, rankings of the RAs in terms of RMSE of monthly 654 total AOD at all the AERONET sites are displayed in Figure 8. It shows that there are cases in that 655 individual RA ranks first over some regions. For example, CAMSRA ranks relatively better than 656 others in South and Southeast Asia. MERRA2 ranks better over North Africa and Arabia 657 658 Peninsula, NAAPS-RA ranks better over North America and Europe while JRAero performs relatively better over Southern North America and the Caribbean. Individual reanalysisRA has 659 mixed results for sites in other regions. AOD RMSE of the MRC is not always the lowest for a 660 given site, but it is relatively low and stable over the globe. This is consistent with the regional 661 662 RMSE result (Figure 6). The consensus wins because of its averaging of independent models. This 663 is consistent with our findings with the ICAP models- (Sessions et al., 2015; Xian et al., 2019). 664

Challenging sites for these RAs are found as marked by the magenta color in Figure 8. These sites 665 exhibit an  $r^2$  value of less than 0.25, and are associated with relatively large AOD bias and/or 666 667 RMSE. Often, when a challenge occurs, it is a common challenge to all models, and no specific 668 model is much better than the others. Some of the causes for the challenges include lack or large 669 uncertainty in local emissions (e.g. Modena in Northern Italy, Mainz in Germany, Cario\_EMA in Egypt, Trelew and CEILAP-RG sites in Argentina), and/or topographic effects that are not 670 resolved in these RAs due mostly to coarse model spatial resolutions (e.g., Mauna\_Loa), and sites 671 that are impacted by mixed pollution and dust (Dushanbe in Tajikistan). 672

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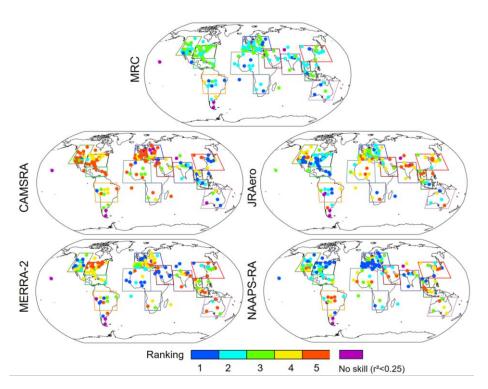


Figure 8: Ranking of aerosol RAs in terms of RMSE of monthly total AOD at 550nm over all the
AERONET sites. Rectangles are used to delineate regions for regional validation, as depicted in
Figures 5, 6, 7. A lower RMSE indicates better performance, with a ranking of 1 being the most
desirable. AERONET sites with a coefficient of determination (r<sup>2</sup>) less than 0.25 are marked in
magenta, indicating a lack of skill from the model.

680

681 Ranking analyses were also conducted on the RMSEs of FM and CM AODs, absolute bias, and 682 <del>coefficient of determination ( $r^2$ )</del> of modal AODs. Figure 9 presents the MRC rankings for all these 683 comparison statistics. In line with the MRC ranking for the total AOD's RMSE, the MRC rankings 684 for other metrics are predominantly ranked first or second, except for the absolute biases, where 685 MRC rankings are often ranked third over North America, South Americas, and Europe for total and FM AODs. For these modes and over these regions, all the RAs have positive biases relative 686 to AERONET. When the biases are in the same sign (positive or negative), it is mathematically 687 natural for MRC to rank in the middle. For CM and FM AODs, there are more sites with  $r^2 < 0.25$ 688 compared to the total AOD. These sites mostly have small values of CM or FM AODs, and reside 689 in regions of opposite-mode dominance, such as FM in Saharan region, CM in northern Europe 690 and N. America. From another perspective, the MRC ranking with respect to correlations is 691 superior to RMSE and then absolute bias. That is, the MRC better captures aerosol variance than 692 the individual models, but is nevertheless subject to overall model biases. The MRC ranking for 693 CM AOD is slightly superior to that of total AOD and then FM AOD. While the MRC ranking is 694

not consistently at the top for a given site or region, it is relatively high and stable, ranking firstfor the global average. No individual RAs could compete with the MRC in that sense.

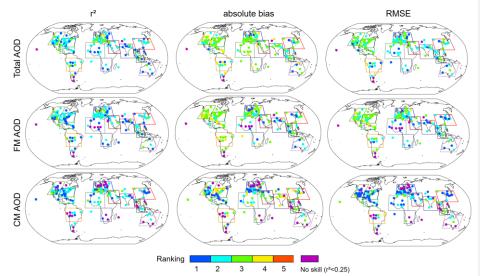


Figure 9. Ranking of the MRC among all the RAs in terms of  $r^2$ , absolute bias, and RMSE of the total, FM, and CM AODs over AERONET sites.

700

In Section 3.1 we depict the spatial distribution of total AODs from all the RAs across the four 702 seasons. In this section, we provide monthly time series of AOD and AOD interannual variabilities 703 for 16 regions (Fig. 10), along with the contributions of speciated AOD to the total AOD for these 704 705 regions- for four seasons and the annual-mean. All the RAs exhibit a similar seasonality and 706 interannual variability of total AOD for all regions, except for the Antarctic and Arctic, particularly 707 during their winter seasons. This disparity arises from the absence of passive satellite AOD data 708 during polar winter, which limits the effect of data assimilation on model AOD (see Xian et al., 2022 for the Arctic region). Even during polar summer, AOD retrievals are often unavailable due 709 710 to high reflectance from surface ice/snow. The polar regions demonstrate the most significant diversity among the RAs in the seasonal cycle and speciation of AOD. The total AOD in JRAero 711 exhibits exceptionally high levels, primarily attributed to elevated sea salt and sulfate AODs (Fig. 712 S5). This anomaly stems from the MASINGAR model used to produce JRAero, which tended to 713 underestimate the removal of aerosols via cumulus convection. Consequently, this led to an 714 715 overestimation of aerosol concentrations in the polar regions and the upper atmosphere. The underestimation of the removal process has been resolved in the current MASINGAR model and 716 the overestimation of AOD over the polar regions will be improved with the JRAero version 717 upgrade. Nevertheless, the polar regions demonstrate the most significant divergence among the 718 719 RAs in the seasonal cycle and speciation of AOD.

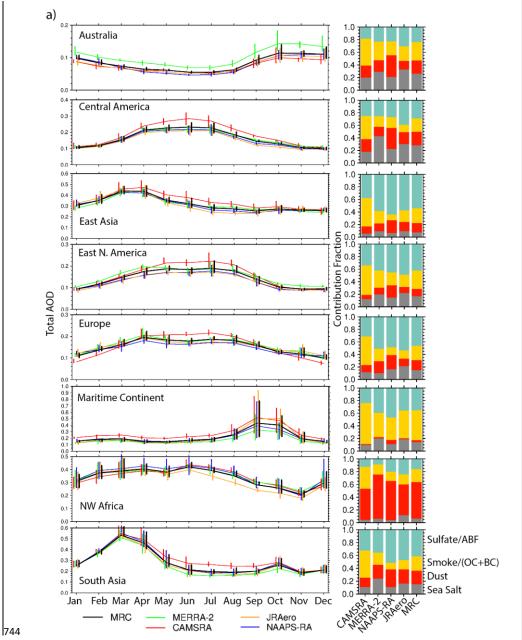
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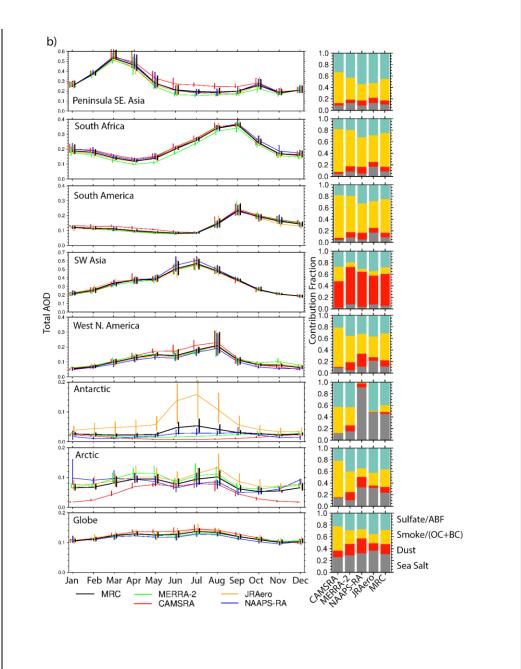
721 The regions that are dominated by BB-smoke, including South Africa, South America, Maritime 722 Continent, Peninsula SE Asia, and western North America, exhibit consistent peak seasons of total

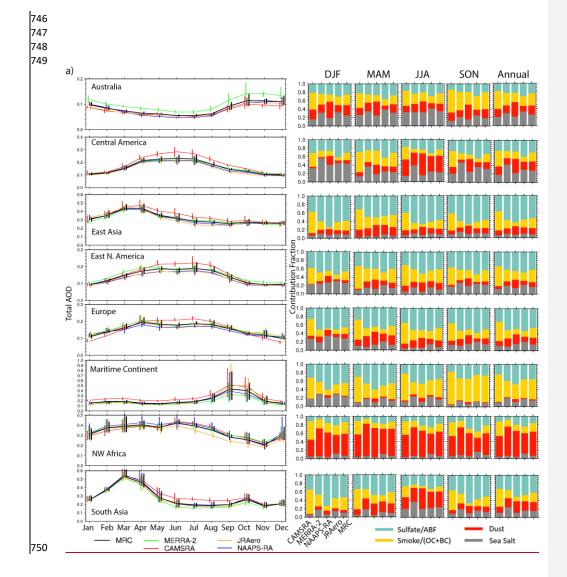
<sup>701 3.4</sup> Seasonality of Regional AODs

AOD with their respective burning seasons. The Maritime Continent and Peninsula SE Asia 723 experience extremely large interannual variations of peak monthly AOD, owing to a strong 724 positive correlation between burning activities and El Nino cycles (e.g., Reid et al., 2012; Xian et 725 al., 2013). The contributions of sulfate/ABF AOD induced by pollution are dominant in East Asia 726 and South Asia, while other aerosol species also make a significant contribution to the total AOD. 727 In Europe and East N. America, sulfate/ABF is also the dominant species; however, the monthly 728 total AOD values are much smaller. All the RAs capture the dominance of dust species in 729 summertime over SW Asia and NW Africa. The relatively high AOD in springtime in NW Africa 730 is partially due to BB in Sahel. In Australia, the peak AOD in Oct-Dec is associated with BB 731 smoke. In Central America, the relatively high AOD in the springtime results from BB smoke. 732 Although quite diverse in AOD magnitude, all RAs tend to have a summertime total AOD peak 733 attributed to dust. For the global average, sea salt AOD has a significant contribution to the total 734 735 AOD as the area of the ocean overwhelms the area of land. Monthly time series of the speciated 736 AODs for all the regions are available in the FigureFig. S5. Overall, the seasonality and interannual 737 variability of total AOD for most regions is very similar among the RAs. Moreover, all RAs have 738 the same dominant species for most regions, but the contributions from different species can be quite different in these RAs. This is a result of the fact that total AOD is constrained within these 739 740 RAs through data assimilation, while speciated AODs are not. Aerosol speciation and the contribution of each species to the total AOD are determined by the construction of the aerosol 741 forecast models, which are very independent in these RAs. 742

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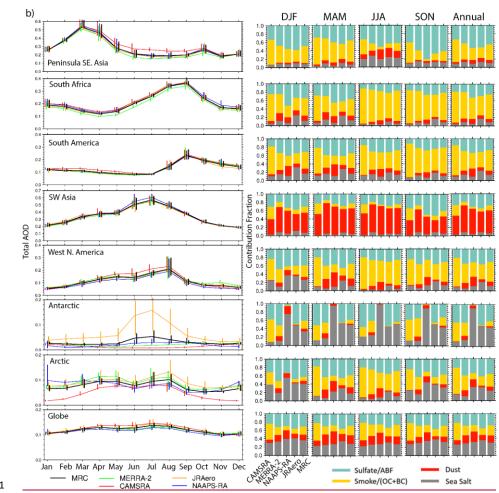


Figure 10. Climatological seasonal cycle of regional mean total AOD (left), and <u>the</u> contribution fraction of speciated AOD to the total AOD for the corresponding regions <u>and seasons</u> from the four RAs and the MRC (right). <u>Bars inIn</u> the seasonal cycle plots <u>represent</u>, <u>bars denote</u> the interquartile range of monthly-mean AOD, <u>illustrating interannual variabilities</u> for <u>the period</u> 2011-2019.

### 757 758

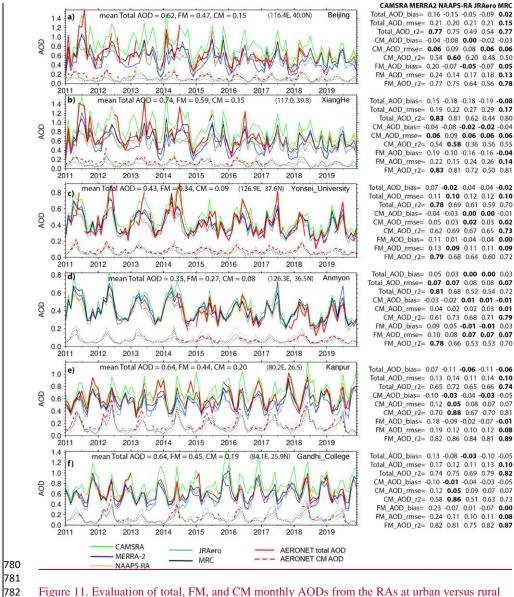
# 3.5 Urban versus Rural areas

759 <u>To evaluate the RAs for urban versus rural areas, three paired sites were selected. Beijing</u>

760 (China), Yonsei University (South Korea) and Kanpur (India) represent urban areas, while their

- 761 corresponding rural areas are represented by the Xiang He, Anmyon and Gandhi College sites
- 762 among the available AERONET sites. Fig. 11 shows the monthly time series of modal AODs
- 763 from the RAs and the MRC, along with their validation statistics against AERONET data. The

764 dominant aerosol mode is FM at all these sites, due mostly to pollution. These sites are also 765 subject to the influence of dust storms in springtime, which contributes to CM AOD. The modal 766 AODs from the four RAs and the MRC generally follow these of AERONET seasonally. The 767 spread among the RAs is relatively large for the Chinese and Indian sites. The spread is relative 768 small for the Korean sites, with the spread being slightly less for the rural site Anmyon than for 769 its corresponding urban site Yonsei University. Regarding bias, RMSE, and r<sup>2</sup>, there is no 770 significant difference between the urban and the corresponding rural sites for each RA and the 771 MRC, despite that  $r^2$  of total AOD tending to be higher for the rural sites than the urban sites. 772 The  $r^2$  of FM AOD also tends to be higher than that of the CM. The RAs and the MRC also 773 capture the decreasing AOD trend in the latter half of the 2011-2019 time period for the Chinese 774 and Korean sites. A more detailed trend analysis will be provided in a companion paper. For the 775 ranking of all RAs in terms of bias, RMSE and  $r^2$ , each individual RA has a few first rankings. 776 MERRA-2 is especially better compared to other RAs at CM/dust AOD for the Indian sites. But 777 in terms of the number of ranking first, the MRC is the winner for all the sites (at least having 5 778 out of 9 statistical variables ranking first for each site). 779



0.02

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32 783 AERONET sites. Sites a), c) and e) represent urban locations in China, Korea and India

- 784 respectively, while sites b), d) and f) denote their corresponding rural sites. Mean Total, FM, and
- 785 CM AODs from AERONET data are presented in the upper panels of the time series plots for
- 786 each site. The right column displays verification statistics for the four RAs and the MRC,

 $\frac{\text{including bias, RMSE and } r^2. \text{ Values in bold indicate the lowest bias or RMSE, or the highest } r^2, \\ \frac{\text{signifying the best ranking among all the RAs.}}{\text{signifying the best ranking among all the RAs.}}$ 

789 790

## 791 **4.** Conclusions

This study compares the monthly average total, and speciated aerosol optical depths (AODs) from 792 four different aerosol reanalyses (RAs). These include the Copernicus Atmosphere Monitoring 793 794 Service ReAnalysis (CAMSRA) developed by Copernicus/ECMWF; the Japanese Reanalysis for Aerosol (JRAero) developed at the Japan Meteorological Agency (JMA); the Modern-Era 795 Retrospective Analysis for Research and Applications, version 2 (MERRA-2) developed by 796 NASA; and the Navy Aerosol Analysis and Prediction System reanalysis (NAAPS-RA); version 797 798 1, developed by the U.S. Naval Research Laboratory. The consensus of the four RAs is also 799 developed for intercomparison. The AODs from these RAs are evaluated with AEROsol Robotic NETwork (AERONET) and the combined-MODIS Dark Target/Deep Blue retrievals (Levy et al., 800 2013; Sayer et al. (2014)) using data from 2011-2019. The following are the conclusions drawn 801 802 from this study:

- Global distribution and magnitude of total AOD demonstrate a high level of similarity among all four RAs. The spread of total AOD among the RAs is small over most regions. Exceptions, where the RAs diverge in total AOD are polar regions and areas affected by specific factors<del>, including that include</del> volcanic outgassing, high terrainsterrain, and certain desert regions.
- 808 2) The relative spread of speciated AODs is considerably larger than that of total AOD. CAMSRA consistently yields higher values for biomass burning (BB) smoke or Organic 809 810 Aerosol (OAMatter (OM) AOD in comparison to other RAs. Meanwhile, NAAPS-RA exhibits generally higher dust AOD values. JRAero has comparatively high biased inland 811 812 sea salt AOD. The diversitydivergence of speciated AODs in regions remote from aerosol sources is large, implying different efficiencies in removal during long-range transport. 813 This phenomenon results from the fact that data assimilation in these RAs constrains total 814 AOD but not speciated AOD. 815
- 3) The seasonality and interannual variability of total AOD in the 16 regions under study,
  with the exception of the Antarctic and Arctic, demonstrate a high degree of similarity
  across the various RAs- and align with the observations. While the dominant species of
  aerosols are consistent across most regions in all RAs, the relative contributions from
  individual species can vary significantly.
- 4) The accuracy of the RAs, as measured by RMSE, bias, and correlation of the total, fine-mode (FM) and coarse-mode (CM) AODs; (i.e. modal AODs), has been verified with AERONET. It is evident that each RA exhibits its own unique regional strengths.
  Specifically, CAMSRA performs better in South and Southeast Asia, MERRA-2 excels in African and Arabian Peninsula dust regions, NAAPS-RA shows relatively better over southern North America and the Caribbean. Common challenges to all the RAs often

include lack or large uncertainty in local emissions, and/or topographic effects, as well as
situations where both FM and CM states are mixed. There is no significant difference in
RAs' performance for urban versus rural areas, despite that rural areas tend to have slightly
higher AOD correlations with observations. RAs show the worst performance in areas
impacted by mixed FM and CM aerosols, such as South Asia and East Asia, and areas that
experience substantial interannual variability in AOD, for instance, Southeast Asia, and the
Maritime Continent. The polar regions present a challenge due to limited observations.

- 835 5) The Multi-Reanalysis-Consensus (MRC), based on an ensemble mean of the four RAs, is not consistently the best performer in terms of RMSE, bias and correlation of modal AODs 836 for a given site or region. However, the MRC generally performs relatively well and 837 838 remains stable, ranking first or second regionally and first globally among all the RAs, especially for correlation and RMSE. The MRC ranking with respect to correlations is 839 superior to RMSE and then absolute bias. The MRC ranking for CM AOD is slightly 840 841 superior to that of total AOD and then FM AOD. The MRC method gains an advantage 842 due to its ability to average independent models.
- 843 The findings presented in this study offer a comprehensive overview of the current state-of-the-art 844 aerosol reanalysesRAs in the context of monthly AOD. The strengths and weaknesses of individual 845 reanalysesRAs and their collective implications will provide valuable information for diverse 846 potential users. Compared to intercomparisons of satellite AOD products, which have shown a 847 typical bias of 15%-25% (which regionally can reach ±50%) and AOD diversitydivergence of 10% 848 over ocean to 100% over certain land areas amongst 14 satellite products (in Schutgens et al., 849 2020), and the intercomparisons of different MODIS products shown in Fig. 1, the biases and 850 diversity divergence of AODs from the four reanalyses RAs are moderate. The MRC product, 851 which is currently a simple ensemble mean of the four RAs, could be potentially improved with 852 regionally-weighted member contributions according to the strengths of the RAs or with aerosol 853 scenario/species-weighted member contributions.

854 The results of the intercomparison highlight areas for improvement in the next generation of 855 aerosol reanalyses<u>RAs</u>. These improvements may include tuning of emission sources and sinks, finer spatiotemporal resolutions, incorporation of additional aerosol species, such as nitrate 856 857 aerosols and dust with different mineralogy, separation of BC and OC from BB emissions in some 858 RAs, and application and enhancement of BB plume rise models. Moreover, some centers are 859 planning to incorporate new observational data, such as OMI Aerosol Index to constrain the amount of absorptive aerosols, which has the potential to enhance simulations of BB smoke and 860 861 dust aerosols (Zhang et al., 2021, Sorenson et al, 2023); Sorenson et al, 2023). Vertical profiles 862 of aerosol backscatter measured by CALIOP and future space-borne lidars may also be 863 incorporated into RAs to help constrain aerosol vertical distribution. Anticipated advancements in emission inventories, retrieval algorithms, space-borne sensors, upcoming satellite missions, and 864 improvements in meteorological and aerosol modelling are expected to drive progress in aerosol 865 866 reanalysesRA.

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# 869 Appendix A: Abbreviations:

- 870 <u>ABF: Anthropogenic and Biogenic Fine aerosols</u>
- 871 <u>AERONET: Aerosol Robotic Network</u>
- 872 <u>AOD: Aerosol Optical Depth</u>
- 873 AVHRR: Advanced Very High Resolution Radiometer
- 874 BB: Biomass Burning
- 875 <u>BC: Black Carbon</u>
- 876 <u>CALIOP: Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP)</u>
- 877 <u>CAMSRA: Copernicus Atmosphere Monitoring System Reanalysis</u>
- 878 <u>CM: Coarse Mode</u>
- 879 <u>FLAMBE: Fire Locating and Modeling of Burning Emissions</u>
- 880 <u>FM: Fine Mode</u>
- 881 ICAP: International Cooperative for Aerosol Predictions
- 882 JRAero: the Japanese Reanalysis for Aerosol
- 883 MASINGAR: Model of Aerosol Species IN the Global AtmospheRe
- 884 MISR: Multi-angle Imaging SpectroRadiometer
- 885 <u>MME: Multi-Model-Ensemble</u>
- 886 MODIS: Moderate Resolution Imaging Spectroradiometer
- 887 MODIS-DT: MODIS Dark Target
- 888 MODIS-DB: MODIS Deep Blue
- 889 MODIS-DA: MODIS data assimilation quality data.
- 890 <u>MRC: Multi-reanalysis-consensus</u>
- 891 <u>NAAPS-RA v1: Naval Aerosol Analysis and Prediction System-Reanalysis version 1.</u>
- 892 MERRA-2 : Modern-Era Retrospective Analysis for Research and Applications version 2
- 893 <u>OM: Organic Matter</u>
- 894 <u>OC: Organic Carbon</u>
- 895 OMI: Ozone Monitoring Instrument (OMI)
- 896 PMAp: Polar Multi-Sensor Aerosol product
- 897 <u>QFED: Quick Fire Emissions Dataset</u>
- 898 <u>RA: ReAnalysis</u>
- 899 <u>RMSE: Root Mean Square Error</u>
- 900 SDA: Spectral Deconvolution Method
- 901 Appendix B: Definition of terminologies
- 902 <u>Root Mean Square Error (RMSE):</u>

903 RMSE = 
$$\sqrt{\frac{1}{n}\sum_{i=1}^{n}(\tau_{model} - \tau_{obs})_{i}^{2}}$$
 where  $\tau$  represents monthly AOD, and n is the total number

- 904 (i.e. month) of observational or model data.
- 905 <u>Bias:</u>  $\tau_{model} \tau_{obs}$

906 <u>Mean error:  $\frac{1}{n} \sum_{i=1}^{n} (\tau_{model} - \tau_{obs})_i$ </u>

907	<u>Mean absolute error:</u> $\frac{1}{n} \sum_{i=1}^{n}  \tau_{model} - \tau_{obs} _i$	
908	<u>Coefficient of determination:</u> $r^2 = \frac{(\sum_{i=1}^{n} (x_i - \overline{x})(y_i - \overline{y}))^2}{\sum_{i=1}^{n} (x_i - \overline{x})(y_i - \overline{y}) \sum_{i=1}^{n} (x_i - \overline{x})(y_i - \overline{y})}$	
909	where $\overline{x}$ and $\overline{y}$ are the mean values of variable x and y.	
910	<u>Multi-Reanalysis-Consensus (MRC):</u> $\frac{1}{m} \sum_{i=1}^{m} x_i$ where <i>m</i> is the total number of the individual	
911	reanalysis, which is 4 for this study.	
912	Spread among the RAs is defined as the standard deviation of all the individual models, ie.,	
913	$\sigma = \sqrt{\frac{1}{m} \sum_{i=1}^{m} (x_i - \overline{x})^2}$ where $x_i$ is individual reanalysis, and $\overline{x}$ is the MRC.	
914	Data Availability	
915 916	All the data supporting the findings of this manuscript can be accessed via the provided links or by requesting them using the contact information provided within those links.	
917	AERONET Version 3 Level 2 data: <u>http://aeronet.gsfc.nasa.gov</u>	
918	MODIS data-assimilation-quality AOD:	
919	https://modaps.modaps.eosdis.nasa.gov/services/about/products/c61-nrt/MCDAODHD.html	
920	CAMSRA AOD: https://www.ecmwf.int/en/research/climate-reanalysis/cams-reanalysis	
921	JRAero product: https://www.riam.kyushu-u.ac.jp/taikai/JRAero/	
922	MERRA-2 AOD:	
923 924	https://disc.gsfc.nasa.gov/datasets/M2TMNXAER_V5.12.4/summary?keywords=%22MERRA- 2%22	
925	NAAPS-RA AOD: https://usgodae.org//cgi-	
925	bin/datalist.pl?dset=nrl_naaps_reanalysis&summary=Go	
927	MRC AOD: https://nrlgodae1.nrlmry.navy.mil/cgi-	
928	<u>bin/datalist.pl?dset=nrl_mre4_post&amp;summary=Go</u>	
929		
930	Supplement	
931		
932	Author contributions	

PX and JSR designed the study. PX performed the data analysis and wrote the paper with
contributions from MA, PRC, KY, TFE, EJH, and JZ on data descriptions and information
collection. All authors contributed to the discussion of the results and revising the paper.

### 936 Competing interests

937 The contact author has declared that none of the authors has any competing interests.

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